^{210}Po in fish from Taboshar uranium mining Pit Lake, Tajikistan.

L. Skipperud, A-G. Jørgensen, B.O. Rosseland, L. S. Heier and B.Salbu

Norwegian University of Life Sciences, Department of Plant and Environmental Sciences, P.O Box 5003, NO-1432 Aas, Norway

Abstract Po-210 in water and different fish species in the Taboshar Pit Lake artificially produced during uranium mining activities in Tajikistan has been determined as part of a Joint project between Norway, Kazakhstan, Kyrgyzstan and Tajikistan. The project aims to assess long term consequences of U mining and tailing legacies, evaluate the need for alternative countermeasures, strengthen the scientific competence and provide support to regulatory authorities in the Central Asian region.

1. INTRODUCTION

In most countries, uranium mining is considered the most hazardous step of nuclear materials production, both in terms of radiation doses and in the number of people affected. Key problems have been associated with the transport of uranium and its daughters in aquatic and terrestrial ecosystems, where radionuclides are transferred from air, water, and soils into plants, fish/animals and finally to man. Special attention is paid to the most hazardous decay products of uranium; ^{210}Pb and ^{210}Po and ^{226}Ra in soils, waters and plants. A certain fraction of uranium and decay products will be present as radioactive particles and the transfer to man occurs via inhalation from resuspension as well as dietary intake. Following the cold war, extensive uranium mining and production took place at numerous sites in Central Asia as part of the nuclear weapon program of the former Soviet Union. The objective of the present work is to provide first hand information on the concentration of the naturally occurring radioactive isotope ^{210}Po, based on speciation analysis of water and by analysis of different fish organs from an area of former uranium mining activity in Tajikistan[1].

2. MATERIALS AND METHODS

Fieldwork was performed in the Republic of Tajikistan in August 2008. The fieldwork focused on the site Taboshar, a mining and tailing area, in particular on samples from the “Pit Lake”, an artificial lake situated in the Taboshar mining area[1].

The Vostokredmet combine was established on the basis of the former Leninabad Mining and Chemical Combine in 1948. During the period of operation about 35 million m^3 of radioactive waste was generated. The waste generated was stored in 9 tailings dumps (a total area of 1 741000 m^2) and in 21 dumps from ores within the mining and processing enterprises [2]. In Taboshar, U rich ores was extracted and a pit lake has been created in the area (figure 1). The 10 m upper part of the Pit Lake is oxic, while the deeper layers are anoxic. A small creek is the only outlet from the lake, maintaining the water level to

about constant. Wild and domestic animals are using the water for drinking purpose.

Sampling of water was performed by *in situ* fractionation, separating radionuclides in terms of size (molecular mass) and charge. Size fractionation was done by using a Millipore membrane filter (mixed cellulose esters) with cut off at 0.45 µm and a Micon Hollow fibre ultra filter having a cut off at 10 kDa [3].
Totally 13 fish (n = 13) of the species Common Goldfish (*Carassius auratus)* from the Taboshar Pit Lake and *C. carpio* and *S. lucioperca* from a reference site (Kayrakkum Reservoir) was sampled. Fish was killed by a blow to the head prior to measurement of length and weight. The dissection procedure followed the EMERGE protocol [4]. Dissected tissues were separately packed in aluminium foil and zipper bags and stored at – 20 ºC. All samples were stored under suitable conditions prior to returning to the Norwegian University of Life Science (UMB) for analysis.


Figure 1. The "Pit Lake" in Taboshar, Tajikistan (Photo B. Salbu)

Sample preparation prior to analyses of ^{210}Po and stable ^{13}C/^{12}C and ^{15}N/^{14}N isotope ratio (identification of food sources and trophic levels of fish) using IRMS were performed in the Isotope Laboratory at UMB. Further, determination of the concentration and distribution of ^{210}Po in liver, bone and muscle of *C. auratus* from study site, and *C. carpio* and *S. lucioperca* from a reference site was performed. Additionally, snails, rush and moss from the area around the study site were analysed for stable carbon and nitrogen isotopes to gain information on a baseline food source and trophic levels of the ecosystem. The fish samples subjected to ^{210}Po analysis included bone (mainly spine), liver (total) and muscle (part). All tissues were analysed separately and related to the individual fish. For total decomposition of solid samples, acid decomposition by UltraCLAVE high performance reactor was used. Chemical separation and thermal plating of ^{210}Po was performed according to the method of Chen *et al* [5]. Po-210 in all samples was determined by alpha spectrometry. Statistical analysis was performed using Minitab. Differences between groups were analysed using ANOVA (general linear model) and $p<0.05$ was set as a criterion for significance.

3. RESULTS AND DISCUSSION

3.1. Water samples

The total activity of 210Po in the Pit Lake is 5.6 ± 0.7 mBq/L (Table 1). According to WHO (2009), the guidance level (GL) of ^{210}Po in drinking water is set at 0.1 Bq/L. In this respect, the activity of Po found in the study site is well below the limit for drinking water.

Table 1. The concentration of ^{210}Po (mBq/L) in water from the Pit Lake, before and after size-, molecular mass and charge fractionation.

Water sample	Po-210 activity concentration mBq/L
Total	5.6 ± 0.7
0.45 μm	5.0 ± 0.5
10 KDa	1.6 ± 0.4
KDa and ion exchange	1.0 ±0.2

The percentile distribution of ^{210}Po, Mn and TOC species are presented in figure 2. The main activity concentration of Po is related to species > 10 kDa with a percentile distribution of particles and colloids being 11 % and 60 %, respectively. The LMM fraction of Po was 28 %, with cations being the major LMM species (62 %).

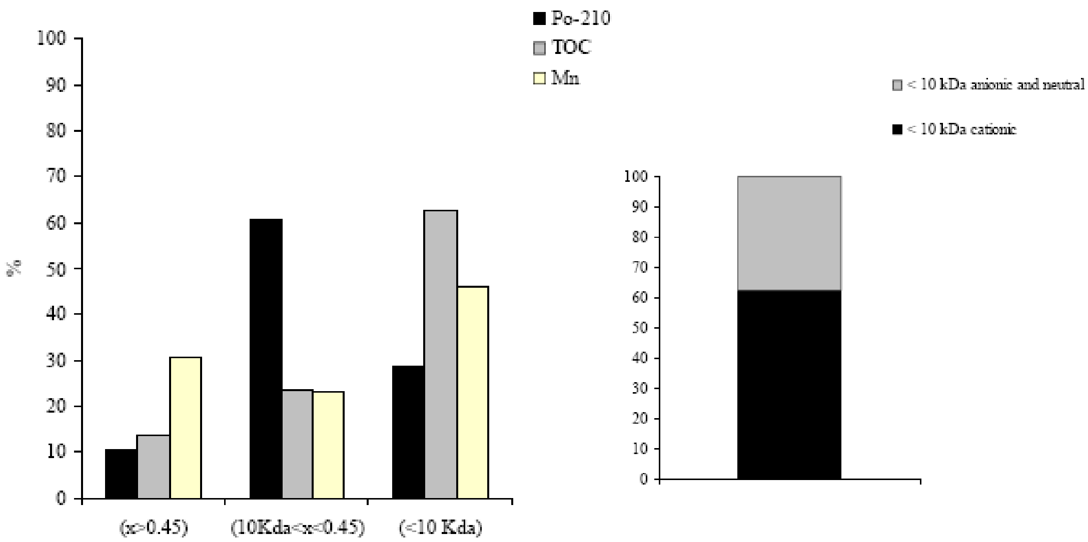


Figure 2. The percentile distribution of ^{210}Po, TOC, and Mn on different cut-off according to size in the Pit Lake; particulate (x > 0.45), colloidal HMM (10 kDa < x < 0.45) and LMM (< 10 KDa) to the left. The percentile distribution of LMM species of ^{210}Po between cation and anionic/neutral species to the right.

3.2. Fish

The activity concentration of ^{210}Po in liver, bone and muscle from all species from the study site and reference site is presented in Table 2. The average and median activity concentration of ^{210}Po in liver, muscle and bone of *C. auratus* was generally higher than the concentration in similar tissues of *C. Carpio* and *S. lucioperca* from the reference site. The highest concentrations of ^{210}Po was in

liver (3673 ± 434 Bq kg^{-1} and 3301 ± 327 Bq kg^{-1}) of *C. auratus*. In the reference site, the highest average and median concentration of ^{210}Po was found in liver of *C. Carpio* (183 Bq kg-1 ± 31 Bq kg-1).

The average and median concentration of ^{210}Po in liver of *C. auratus* from study site was generally higher than the average and median concentration of ^{210}Po in bone and muscle. However, the minimum observation of the concentration of ^{210}Po in liver is lower than the minimum observation in bone and muscle. This observation is considered an outlier as no other individual of *C. auratus* show values < 592 Bq / kg. Additionally, the majority of individuals have a concentration > 3000 Bq kg^{-1}. The activity concentration in liver was significant higher than in muscle and bone ($p=0.017$) of *C. auratus*. There is a distinct difference in the activity concentration of ^{210}Po observed in the liver of *C. carpio* compared to *S. lucioperca* from the reference site. *S. lucioperca* show relatively low activity concentration compared to *C. carpio* also in muscle. However, it is slightly higher in bone. Based on liver and muscle it seem as *C. carpio* has a higher activity concentration of ^{210}Po than *S. lucioperca*.

The activity concentration found in muscle and liver of *C. auratus* from study site and *C. carpio* and *S. lucioperca* from reference site was compared to the activity concentration found in *S. lucioperca* from Kyrgyzstan and Kazakhstan (fig. 3). The average activity concentration of ^{210}Po in muscle tissue of *C. auratus* from the study site increased with a factor of ~250 and ~500 compared to ^{210}Po in muscle of *S. lucioperca* from Kyrgyzstan and Kazakhstan, respectively. Further, the activity concentration of ^{210}Po in *C. auratus* increased with a factor of ~400 compared to the activity concentration in liver of *S. lucioperca* from Kazakhstan.

Table 2. The average (avg.) and median (med) concentration of ^{210}Po (Bq kg^{-1}) (ww) in liver (l), bone (b) and muscle (m) of C. auratus (n = 13) from study site, and C. carpio (n = 2) and S. lucioperca (n = 1) from reference site. The counting error for average and median activity concentration of ^{210}Po is presented in Bq kg^{-1}.

Species (tissue)	**^{210}Po** Avg Bqkg^{1}	**STD** Bqkg^{1}	**Med** Bq kg^{-1}	**Dev** Bq kg^{-1}	**Range** Bq kg^{-1}
C.auratus(l)	3673	± 434	3301	± 327	68-9714
C.auratus(b)	704	± 98	650	± 93	264-1392
C.auratus(m)	409	± 87	287	± 49	128-1279
C. carpio(l)	183	± 31	183	± 31	80-284
C.carpio(m)	14	± 4	14	± 4	4-23
C.carpio(b)	7	± 2	7	± 2	3-11
S.lucioperca(l)	11	± 8	-	-	-
S.lucioperca(b)	8	± 2	-	-	-
S.lucioperca(m)	2	± 1	-	-	-

l = liver, b = bone, m = muscle

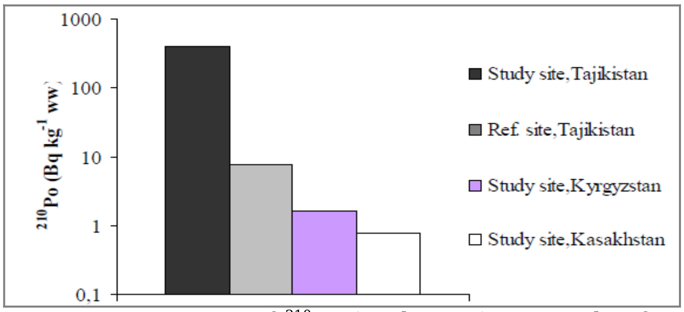


Figure 3. The average concentration of ^{210}Po (Bq/kg ww) in muscle of *C.auratus* from the Pit Lake and *C.carpio* from the Kairrakkum Lake (Ref. site) in Tajikistan, compared to the concentration in *S.lucioperca* from the Issyk-Kyl Lake, Lake Kyrgyzstan and from the Pit Lake, Kurdai, Kazakhstan.

3.3. Throphic level

Figure 4 shows the relationship between stable carbon isotopes ($^{13}C/^{12}C$ = δ13C) and nitrogen isotopes ratios ($^{15}N/^{14}N$ = δ15N) in individual fish, snail, rush and moss from the Pit Lake.

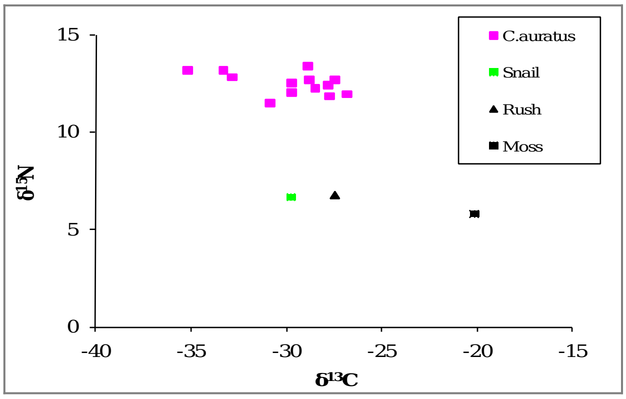


Figure 4. The level of δ^{13}C (carbon source) and δ^{15}N (trophic level) in *C.auratus*, plants and snail from the Pit Lake.

All the *C. auratus* collected in Pit Lake were found to be within the same trophic level. As the golden carp was two tropic levels above the primary producers and snails, the feed was probably zooplankton. In the reference site, the activity concentration in liver of *C. carpio* (182 ± 31 Bq kg^{-1}) was higher than in *S.lucioperca*, although the δ^{15}N showed that *S. lucioperca* represented a higher trophic position.

3.4. Bioconcetration and accumulation of ^{210}Po

The bioconcentration factor (fig.5) for ^{210}Po was calculated for bone, liver and muscle and the results clearly demonstrate high accumulation of ^{210}Po in *C.auratus*, especially in the liver. Linear correlation between ^{210}Po in liver and δ^{15}N were found (Figure 6) which could indicate biomagnification of ^{210}Po. As all the fish collected were found to be representative for the same trophic level, more investigations are needed to investigate any potential biomagnifications properties of ^{210}Po.

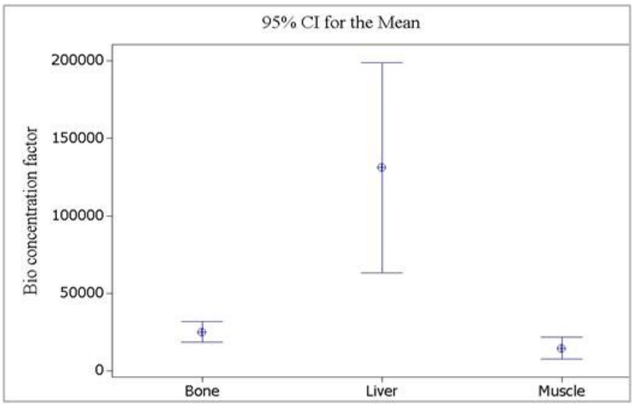
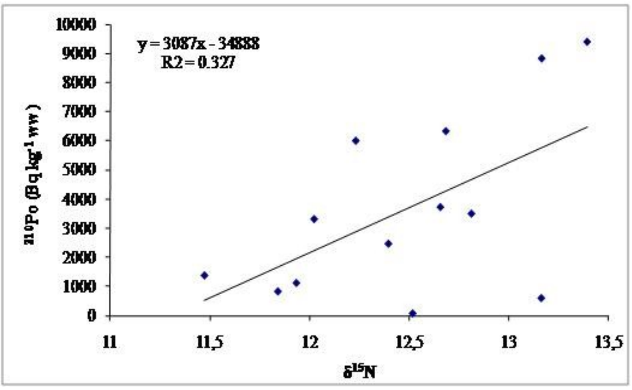


Figure 5. Bioconcentration factors (BCF) of ^{210}Po in bone (2 x 10^4),

liver (10^5) and muscle (10^3) in *C.auratus* collected in Artificial lake (top), and Scatterplot of the relationship between $\delta^{15}N$ and the activity concentration of ^{210}Po in liver of *C.auratus* (bottom).



The concentration of contaminants in organisms often shows interspecific and intraspecific variation. Such variation may be explained by several factors including the route of uptake, age, size, feeding habits and trophic level [6; 7]. Additionally, various contaminants are distributed differently throughout the tissues of organisms due to different mechanisms such as the biochemical characters of the element and specific physiology of tissues [7]. The distribution of ^{210}Po observed in fish species of this study is in line with observations in organisms and fish from other studies performed both in freshwater and marine waters. According to Durand et al [8] it is well known that ^{210}Po is accumulated and that the liver of fish may contain especially high concentrations. This is in line with ideas on general metal uptake in fish, with increased uptake of metals in metabolically active tissues. Durand et al [9] further strengthened the observation of ^{210}Po and its strong affinity to bind to iron containing proteins. They found that ferritin and hemocyanin were molecular traps in the liver tissues of fish.

3.5. Risk assessment

The US Nuclear Regulatory Commission has set the annual limit of intake (ALI) of ^{210}Po through ingestion 5 KBq/year, or approximately 15 Bq/day [10]. An intake of 50g /day of fish from the Pit Lake in Taboshar would result in exceeding the ALI, and considering an average meal of fish (150 -300 g) these levels are alarming. Groups at risk, such as pregnant women and children should not consume fish from the Pit Lake in Taboshar and a high intake of fish from this lake is not recommended for the general population.

4. CONCLUSIONS

The activity concentration of ^{210}Po in tissues of *C. auratus* from study site was generally higher than in similar tissues of *C. carpio* and *S. lucioperca* from the reference site. The activity concentration in liver of *C. carpio* (182 ± 31 Bq kg^{-1}) was higher than in *S.lucioperca*, although the $\delta^{15}N$ showed that *S. lucioperca* represented a higher trophic position. Generally, the highest average and

median activity concentration of ^{210}Po was observed in liver, and the lowest in muscle for all species from both sites. The BCF values observed in the tissues of *C. auratus* was significant higher in liver compared to bone and muscle tissues indicating that ^{210}Po is strongly accumulated in the liver of *C. auratus*.

In regards to the recommended ALI for ^{210}Po, the concentrations of ^{210}Po in muscle tissues of *C. auratus* is alarming, as there is a high probability for the local population to exceed the recommended ALI through consumption of fish from Dead Lake.

Acknowledgements

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